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Temperature-induced pair correlations in clusters and nuclei

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The pair correlations in mesoscopic systems such as *nm*-size superconducting clusters and nuclei are studied at finite temperature for the canonical ensemble of fermions in model spaces with a fixed particle number: i) a degenerate spherical shell (strong coupling limit), ii) an equidistantly spaced deformed shell (weak coupling limit). It is shown that after the destruction of the pair correlations at $T = 0$ by a strong magnetic field or rapid rotation, heating can bring them back. This phenomenon is a consequence of the fixed number of fermions in the canonical ensemble.

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The pair correlations in a macroscopic superconductor are destroyed by increasing the temperature or the external magnetic field. The critical field which marks the boundary between the superconducting and normal phases, is a decreasing function of the temperature T. The BCS theory, which is the mean field approximation based on the grand canonical ensemble, describes very accurately this regime. Applying the grand canonical mean field approach to rotating nuclei [1], where the angular velocity plays the role of the magnetic field, gives an analogous result: The angular velocity where the pair correlations disappear decreases with the temperature. Nuclei and atomic nano-size clusters are composed of a small and fixed number of particles, the single-particle spectrum is discrete and the level spacing is comparable with the pair gap. Due to these facts, the fluctuations of the order parameter become important, which smear out the transition from superconducting to normal phase [1, 2, 3, 4, 5] and lead to pronounced differences between a system with even and odd particle number [6].

In order to properly take into account these fluctuations one has to use the canonical ensemble. The most direct way is to calculate the partition function from the exact eigenvalues of the Hamiltonian, which is possible for some models. In this Letter we study the simple model of fermions occupying an isolated shell of single particle states and interacting by a pairing force. We shall demonstrate that at zero temperature the magnetic field or the angular velocity attenuate the paring in a step-wise manner until it disappears completely above a critical value. *For a mesoscopic system in the strong magnetic field heating may bring back the pair correlations.* This surprising effect is a consequence of the fixed number of fermions in such a small system. The reduction of the fluctuations in particle number leads to a strong increase of the fluctuations of the order parameter, which constitute the pair correlations above the critical field. A re-entrance of pair correlations has first been discussed by Balian, Flocard and Veneroni [6], who studied ensembles with either only even numbers of particles or only odd numbers of particles. We shall demonstrate that the more stringent restriction to a fixed number of particles, which is realized in small systems, leads to qualitatively different results.

First we consider fermions in a spherical potential. Then the electronic states in the cluster (the spin-orbit coupling can be neglected) are characterized by l, the orbital momentum of the shell, its z-projection λ and spin projection σ, which we label by $k = \{\lambda, \sigma\}$. The condition, $k > 0$, used below in Eq. (1), means that $\lambda + \sigma > 0$. Due to the strong spin-orbit splitting in nuclei the nucleonic states in a shell are specified by the total angular momentum j, ($\vec{j} = \vec{l} + \vec{s}$), and its projection $m \equiv k$. We assume that there are only pair correlations within the last partially filled shell. The corresponding "single-shell" Hamiltonian

$$H = H_{\text{pair}} - \omega M, \quad M = L_z + gS_z, \qquad (1)$$
$$H_{\text{pair}} = -GA^{+}A, \quad A^{+} = \sum_{k>0} a_k^{+} a_{\bar{k}}^{+},$$

consists of the pairing interaction H_{pair}, which acts between the valence fermions in the last shell with the strength G, and the "cranking term". The z-components of the total orbital angular momentum and spin, which are the sums of all valence fermion contributions, are denoted by L_z and S_z. The operator A^{+} creates a fermion pair in the time-reversal states $(k, \bar{k})$. In the case of clusters, the cranking term represents the interaction of the electrons with the magnetic-field, where we introduce the Larmour frequency $\omega = \mu_B B$ and the Bohr magneton μ_B. The gyromagnetic ratio $g = 2$. In the case of nuclei, the cranking term takes into account the rotational perturbations at the angular frequency ω and $g = 1$.

The eigenvalues of $H_{\text{pair}}(\omega = 0)$ are [7]

$$E_\nu = -\frac{G}{4}(N_{\text{sh}} - \nu)(\Omega + 2 - N_{\text{sh}} - \nu). \qquad (2)$$

were N_{sh} is the number of particles in the shell, which consists of Ω degenerated single-particle states and $\Omega = 4l + 2$ for clusters and $2j + 1$ for nuclei. The seniority ν, which is the number of unpaired particles, is constrained by $0 \leq \nu \leq N_{\rm sh}$. We assume that $\nu \leq \Omega/2$, otherwise one can pass to the hole representation. Each ν-state at $\nu \geq 1$ is degenerated. The eigenvalues of the Hamiltonian (1) are

$$E_{\nu,i}(\omega) = E_\nu - \omega M_{\nu,i}. \tag{3}$$

The index i involves additional quantum numbers of the ν-state including the spin and possible orbital momenta and their z-projections for clusters or the total angular momentum of ν nucleons and its projection for nuclei. In the case of clusters, $\mu_B M_{\nu,i}$ is the total magnetic moment of ν particles while in nuclei $M_{\nu,i}$ is the angular momentum z projection of ν nucleons. For simplicity, we refer to M as the angular momentum and omit the index i for the states with minimal energy at each ν, which have maximal M_ν.

The canonical partition function Z and the mean value $\bar{M}$ are given by

$$Z(T,\omega) = \sum_{\nu,i} \exp(-\frac{E_{\nu,i}(\omega)}{T}), \tag{4}$$

$$\bar{M}(T,\omega) = \frac{T}{Z}\frac{\partial Z}{\partial \omega}. \tag{5}$$

The evaluation uses the fact that for each ν the sum over i can be reduced to a sum over single-particle projections. For nuclei, the numerical diagonalization procedure described in Ref. [8] is used, which permits us also to treat a non-degenerate shell (see below). We consider only the part of $\bar{M}$ which is generated by the particles near the Fermi surface that participate in the pair correlation. The contributions for the other particles can be found in ref. [9] and will be discussed together with the details of the evaluation of the sums for clusters in an forthcoming extended paper [10].

We introduce the "canonical" pair gap $\Delta_{\rm can}$ as a measure of the correlation energy

$$\Delta^2_{\rm can}(T,\omega) = \frac{G}{Z(G)}\sum_{\nu,i}\langle \nu,i|A^+A|\nu,i\rangle \exp(-\frac{E_{\nu,i}(G)}{T})$$
$$-\frac{G}{Z(G=0)}\sum_{\nu,i}\langle \nu,i,0|A^+A|\nu,i,0\rangle \exp(-\frac{E_{\nu,i}(G=0)}{T}), \tag{6}$$

where $|\nu,i,0\rangle$ denote the uncorrelated fermion configurations in the shell. The second term subtracts the expectation value of the pairing interaction in an ensemble of uncorrelated fermions. A detailed discussion of the proper definition of $\Delta_{\rm can}$ was given in the review [3]. Applying the mean-field approximation and the grand canonical ensemble to our model, the thus introduced $\Delta_{\rm can}$ becomes the familiar BCS gap parameter $\Delta_{\rm mf}$. However, $\Delta_{\rm can}$ must be clearly distinguished from $\Delta_{\rm mf}$ because it incorporates the correlations caused by the fluctuations of the order parameter. We take as energy scale $E(0) = G\Omega/4$, the quasiparticle energy at $T = \omega = 0$. A value of $E(0) = 0.3 - 0.4$ meV was found for Al-clusters with radii $R = 5 - 10$ nm [11], which sets

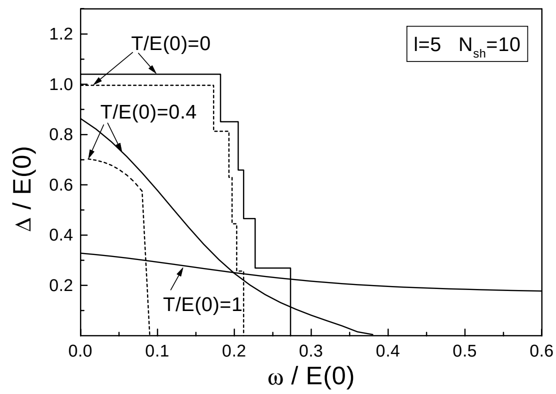

FIG. 1: Canonical gap $\Delta_{\rm can}(T,\omega)$ (full lines) and the mean-field gap $\Delta_{\rm mf}(T,\omega)$ (dotted lines) v.s. the frequency ω for a spherical shell.

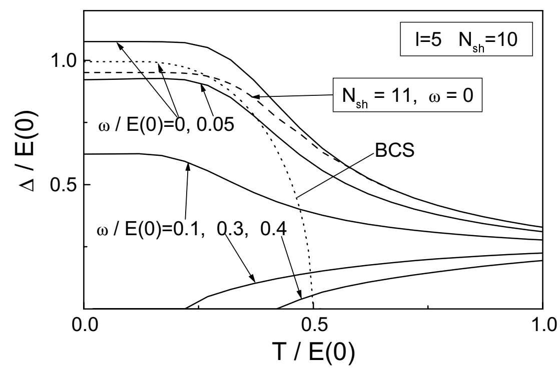

FIG. 2: Canonical gap $\Delta_{\rm can}(T,\omega)$ for even (full lines) and odd (the dashed line) particle number, and the mean-field gap $\Delta_{\rm mf}(T,\omega)$ (dotted line -BCS) v.s. the temperature T for a spherical shell.

the energy scale. The nuclear mass measurements give $E(0) = 0.8 - 1.5$ MeV. Let us first consider the destruction of the pair correlations at $T = 0$. Since M_ν increases with ν, the energy $E_\nu(\omega)$ becomes smaller than $E_{\nu-2}(\omega)$ at the frequency

$$\omega_\nu(M_\nu - M_{\nu-2}) = E_\nu - E_{\nu-2}. \tag{7}$$

When the state of the lowest energy changes from $\nu - 2$ to ν, two more electron states are blocked and the pair correlations are reduced. The last step at the critical frequency $\omega_{\rm crit}$ leads to the maximal seniority $\nu_{\rm max}$, where all particle states are blocked and the pairing is completely destroyed. Fig. 1 illustrates the step-wise destruction of pairing by blocking for a half-filled $l = 5$ shell, which is the Fermi level in a spherical Al-cluster with about 10^3 atoms.

Fig. 1 also shows the gap $\Delta_{\rm mf}(\omega,T)$ obtained by applying the mean-field approximation (cf. [7, 9]) to the single-shell model. For $T = 0$, the pair correlations are more rapidly destroyed than for the exact solution. The quantal fluctuations of the order parameter generate additional pair correlations. For finite T, the mean-field gap behaves as known from macroscopic superconductors: $\Delta_{\rm mf}(\omega, T)$ is reduced at $T = 0.4E(0)$ and reaches 0 at a lower value of ω. This is the familiar shift of the critical frequency $\omega_{\rm crit}$ toward smaller values with increasing T. However, the canonical gap $\Delta_{\rm can}(\omega, T)$ behaves differently: The abrupt drop around $\omega_{\rm crit}$ is smoothed out by the fluctuations of the order parameter. Moreover *substantial pair correlations appear in the region above $\omega_{\rm crit}$, which increase with T*. The comparison with $\Delta_{\rm mf}(\omega, T)$ shows that the fluctuations contribute more at finite T than at $T = 0$. For $T = E(0)$, the pair correlations fall off very gradually with ω. The nuclear case is quite similar.

Fig. 2 shows how these temperature-induced pair correlations appear with increasing T. For $\omega = 0$, there is a pronounced drop of $\Delta_{\rm can}$ around $T_{\rm crit}(\omega = 0) = E(0)/2$, where the mean-field gap $\Delta_{\rm mf}$ goes to zero. Above this temperature there is a long tail of pair correlations caused by the fluctuations. For $\omega = 0.05\ E(0)$, the drop is shifted to smaller T by about the same amount as the $T_{\rm crit}$ of the mean-field solution (not shown). This trend continues for $\omega = 0.1\ E(0)$. For larger $\omega \geq \omega_{\rm crit}$, *the pair-correlations built up with increasing T*.

The temperature-induced pairing can be understood in the following way. At $T = 0$, all electrons are unpaired when the state of the maximum seniority becomes the ground state for $\omega > \omega_{\rm crit}$. At $T > 0$, excited states with lower seniorities enter the canonical ensemble, which reintroduce the pair correlations.

The degenerate spherical shell corresponds to the strong coupling limit ($\Delta/d \gg 1$, d distance between the levels) of ref. [6]. For $\omega = 0$, one may compare $\Delta_{\rm can}$ of the present work with Δ_F obtained for the ensemble with good particle number parity in ref. [6], which we will refer to as BVF in what follows. For $T < 0.5\ T_{\rm crit}$, both are similar. However for $T > T_{\rm crit}$, $\Delta_F = 0$ whereas $\Delta_{\rm can}$ remains finite up much higher temperatures. The case $\omega \neq 0$ is not shown in ref. [6] for the strong coupling limit.

In order to investigate the consequence of deviations from the spherical symmetry, we used the deformed shell-model described in [8]. The Hamiltonian is given by

$$H = \sum_k e_k a_k^+ a_k + H_{\rm pair} - \omega M. \qquad (8)$$

The solutions are found by numerical diagonalization in the configuration space of a j-shell.

Figs. 3 and 4 illustrate the case of a cluster without spherical symmetry. The half- filled shell consists of 12 equidistant levels, each of which contains two states with spin up and down. Our choice of the level distance $d = 0.85\ E(0)$ corresponds to weak coupling $\Delta \approx d$ and is realistic for the nano-clusters. We assume an irregular cluster shape. As a consequence, the orbital momentum is quenched and only the spins contribute to the magnetic moment $\mu_B M$. The behavior of $\Delta_{\rm can}$ and $\bar{M}(T, \omega)$ is similar to the spherical case. The step-wise destruction of the pair correlations corresponds to subsequent spin flips of electrons, which increase M by 2. The temperature-induced pairing appears in the region above $\omega \sim 2\ d$. The spin flips have been observed in tunneling experiments on Al nano-clusters [3]. The mean-field gap $\Delta_{\rm mf}(T = 0)$ breaks down at the first flip, as discussed by Braun et al. [12] in their analysis of the tunneling spectra based on the mean-field approximation. However, $\Delta_{\rm can}(T = 0)$ is more stable and disappears only after all levels are blocked by subsequent spin flips. Fig. 5 demonstrates that the temperature dependence of $\Delta_{\rm can}$ is qualitatively similar to the strong coupling case in Fig. 2. A new feature is that sometimes $\Delta^{\rm odd}_{\rm can} > \Delta^{\rm even}_{\rm can}$. For the curves $\omega = 2d$ in Fig. 5, this happens because the frequency is above the first crossing in the even system (cf. Fig. 3) but still below the first crossing in the odd system, which

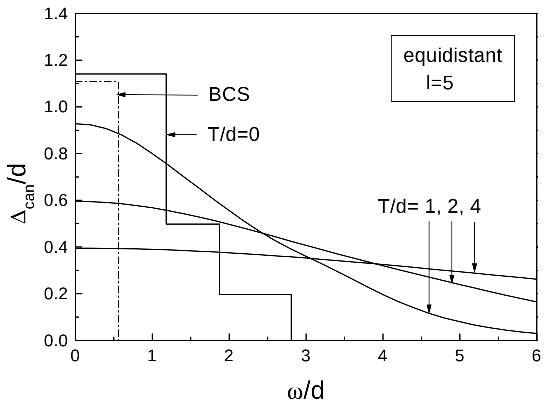

FIG. 3: Canonical gap $\Delta_{\rm can}(T, \omega)$ for an equidistantly spaced $l = 5$ shell in a cluster. The mean-field gap $\Delta_{\rm mf}(T, \omega = 0)$ is shown by the dash-dotted line (BCS).

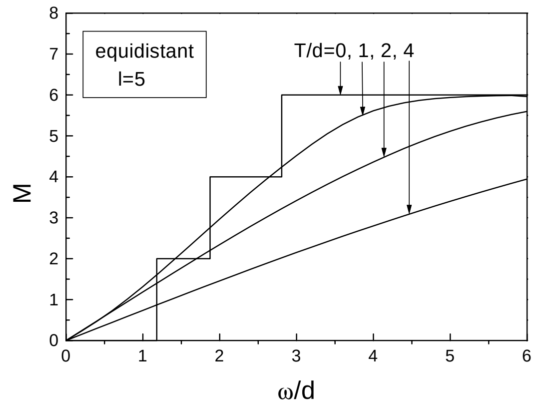

FIG. 4: Angular-momentum $\bar{M}(T, \omega)$ for an equidistantly spaced $l = 5$ shell in a cluster.

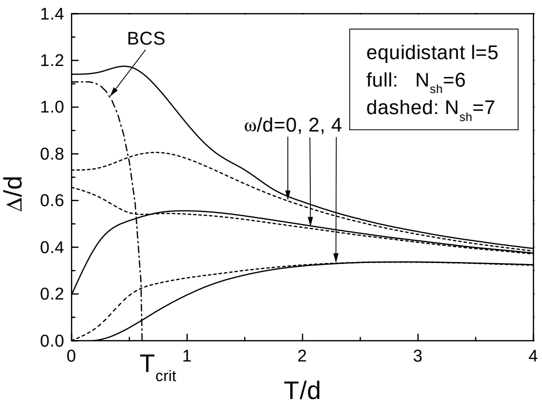

FIG. 5: Canonical gap $\Delta_{\rm can}(T,\omega)$ for an equidistantly spaced $l=5$ shell in a cluster with even (full line) and odd (dotted line) particle number. The mean-field gap $\Delta_{\rm mf}(T,\omega=0)$ is shown by the dash-dotted line (BCS).

means two states are blocked in the even but only one in the odd system. For $\omega=4d$ the analogous happens at the third crossing. In the region of high temperatures ($T>2T_{\rm crit}$) the canonical pairing gaps for odd and even systems practically coincide because all members of Z contribute with a similar weight. At small T they differ because only the states with small seniorities are important.

Our choice of parameters lies between to the weak coupling cases $\Delta/d=1.14$ and 1.19 of the grand canonical ensembles with even or odd particle number studied by BVF ($\Delta/d=w_F\Delta$ in [6]). As in the case of strong coupling, $\Delta_{\rm can}$ decreases very gradually for $T>T_{\rm crit}$, whereas Δ_F of BVF drops sharply to zero at $T_{\rm crit}$. However, also in the region $T<T_{\rm crit}$ the results of the two approaches are different. The differences between the even and odd systems are much less pronounced in the canonical ensemble. For $\omega=0$, $\Delta/d=1.14$, and odd particle number, BVF find $\Delta_F=0$ for $T<0.2\,T_{\rm crit}$, $\Delta_F\neq 0$ for $0.2\,T_{\rm crit}<T<T_{\rm crit}$, and $\Delta_F=0$ for $T>T_{\rm crit}$, which they called the re-entrance phenomenon of pairing. As seen in Fig. 5, the strong fluctuations in the canonical ensemble keep $\Delta_{\rm can}$ finite in the whole temperature interval, i. e. there is no re-entrance of pairing for $\omega=0$ and odd particle number. On the other hand, the canonical ensemble gives temperature-induced pair correlations for $\omega>2\,d$ both for even and odd particle number, which has not been found for grand-canonical ensembles with fixed the particle number parity. It should be mentioned that BVF considered a systems with $N=100$, 101 whereas we studied systems with $N\sim$ 10, for which the conservation of particle number is more important.

We have also studied the case of a half-filled $j=11/2$ shell in a deformed axial nucleus by assuming that e_k in Eq. (8) is proportional to k^2. We chose the distance $e_{7/2}-e_{5/2}=0.28\;E(0)$, which is realistic. If the axis of rotation is parallel to the symmetry axis, the behavior is similar to the deformed clusters, except the steps in M are different from 2. If the axis of rotation is perpendicular to the symmetry axis, the projection M of the angular momentum is no longer conserved. Then $\bar{M}(T=0,\omega)$ is no longer a step function and $\Delta_{\rm can}(\omega,T=0)$ decreases in a gradual manner. The increase of $\Delta_{\rm can}(\omega,T)$ with T is found to be weak. Hence, temperature-induced pairing is expected in nuclei that build up large angular-momentum by aligning the individual angular-momenta of the nucleons near the Fermi surface. These are either spherical nuclei (see Figs. 1,2) or the high-K isomers (see e.g. [13]).

In summary, at very low temperature an increasing external magnetic field causes the magnetic moment of small superconducting clusters ($R<5\;nm$) to grow in a step-wise manner. Each step reduces the pair correlations until they are destroyed. However, with increasing temperature the steps are washed out and substantial pair correlations re-appear for high field strengths, where they are quenched at $T=0$. Nuclei that built up angular momentum along a symmetry axis behave in an analogous manner. The pair correlations are destroyed in a step-wise manner by subsequent alignment of the angular momenta of individual nucleonic orbitals with the symmetry axis. These steps are washed out with increasing temperature and pair correlations appear at values of the rotational frequency, where they are quenched at $T=0$. This phenomenon of temperature-induced pairing reflects the strong fluctuations of the order parameter in very small systems with a fixed particle number.

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